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Characterization of indium iodide detectors for scintillation studies

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Abstract

Indium iodide (InI) is a wide bandgap semiconductor ($E_g = 2.0$ eV) and has been investigated as an optical detector material for use in γ-ray scintillation spectroscopy. Single crystals of InI have been grown by the Bridgman process using zone-refined starting material and optical detectors have been fabricated from such crystals. The performance of these detectors has been investigated by measuring their quantum efficiency, direct X-ray detection characteristics, and electrical resistivity. The InI photodetectors have been coupled to CsI(Tl) scintillators and room-temperature energy resolutions of 7.5% (FWHM) and 9.8% (FWHM) were recorded for 662 keV and 511 keV γ-rays, respectively. Successful γ-ray detection has also been accomplished with InI photodetectors coupled to LSO (Lu_2SiO_5:Ce) scintillators, and a resolution of 14% (FWHM) has been recorded for 511 keV γ-rays. Finally, analysis of the electronic noise behavior of the InI detectors has been performed.

1. Introduction

InI is a wide bandgap semiconductor ($E_g = 2.0$ eV) with base-centered orthorhombic crystal structure, belonging to the D_{2h}^{17} space group [1]. It melts at a relatively low temperature (351°C), and does not exhibit any solid–solid phase transition. Hence, crystals of InI can be grown by melt-based processes (such as the Bridgman technique) which are generally easier, faster and more economical. Prior investigation of InI optical detectors has shown that these detectors have very good quantum efficiency ($\approx 60\%$) in the 400 to 700 nm wavelength region [2] where most inorganic scintillators emit. In addition, InI crystals have exhibited high resistivity ($> 10^{10}$ Ω cm) and good charge-transport parameters (mobility-lifetime product of electrons $= 7 \times 10^{-5}$ cm^2/V), even when no starting material purification was performed [2]. InI detectors have also been successfully operated at elevated temperatures [3]. Hence, InI crystals are quite promising for scintillation spectroscopy.

A more detailed evaluation of InI photodetectors has been performed recently and is reported in this article. This investigation involved the purification of commercially available InI by zone refining, crystal growth of InI using purified starting material, fabrication of optical detectors from such crystals, and, finally, characterization of electrical, optical, γ-ray detection, and noise properties of InI photodetectors.

2. Photodetector fabrication

In order to produce ultrapure InI crystals for photodetection experiments, commercially available InI material (99.999% from APL, IL) was purified using the zone-refining process. A computer controlled, 2-zone furnace in a horizontal configuration was used and about 50 zone-refining passes were carried out using evacuated quartz ampoules as crucibles. The design and operation of our zone refiner used for purifying similar materials such as lead iodide and thallium bromide was described in detail earlier [4,5], and indium iodide purification was carried out in a similar manner. Zone-refined indium iodide appeared transparent on the pure end, while the impure end was opaque and dark (due to accumulation of impurities). Small sections of InI material from the pure end were then used for Bridgman crystal growth experiments during which the purified material was sealed in an evacuated quartz ampoule and dropped slowly (5 mm/day) through a vertical furnace. The furnace temperature was set at 450°C which was high enough to melt the InI charge in the ampoule. The material crystallized as the ampoule approached the bottom of the furnace due to the sharp temperature gradient at that point. Indium iodide crystals with a 1.4 cm diameter and 5 cm long were grown in this

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manner. The crystals were transparent and appeared dark red in color as can be expected from the InI bandgap of 2.0 eV.

These crystals were cut into slices (typically 0.7–1 mm thick) using a wire saw. The slices were lapped and polished using alumina slurry, and then etched using 5% bromine in a methanol solution. An alternative etchant, dilute nitric acid solution, was also tried and similar results (as bromine-methanol etch) were observed. The resulting InI slices were about 500 μm thick and photodetectors were fabricated by depositing electrodes on them. The top, optically transparent electrode was fabricated by evaporating very thin (≤ 70 Å) gold films (which show excellent optical transmission properties in the visible and near-UV region) on the InI crystal surface. The other electrode was fabricated by applying a graphite layer on the back face, and wires were connected to both electrodes. The detectors fabricated in this manner were secured on ceramic substrates. Indium crystals have shown some moisture sensitivity in the past [3] and hence some detectors were encapsulated in an organic coating-Parylene C which was applied by evaporation at SSC, Nashua, NH. Parylene C has been used successfully as an encapsulant on other detector materials such as silicon and HgI_2, and our initial results indicate that it also works well with InI detectors.

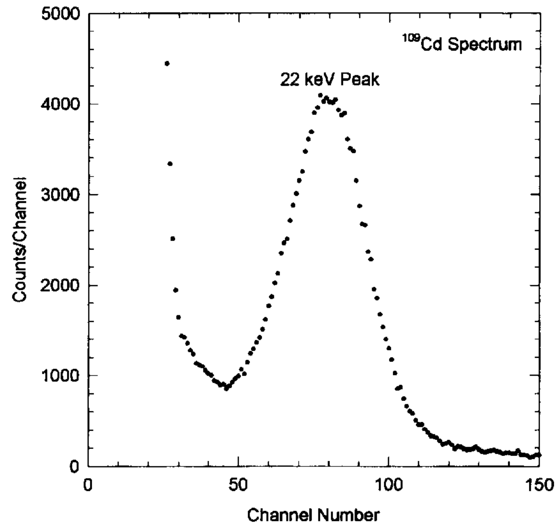

Fig. 1. Direct detection of X-rays using an InI detector. The pulse height spectrum shows a ^{109}Cd spectra with a 22 keV photopeak, recorded with an InI detector (5×5 mm^2 in area and 500 μm thick) operating at 250 V bias and 2 μs integration time.

3. Electrical, semiconducting and optical properties

Electrical properties such as the resistivity and the charge-transport properties of InI crystals were evaluated. The resistivity was measured by recording the current versus voltage (I–V) characteristics of the InI detectors. The I–V behavior was linear and from the slope of the linear regression fit to the data, the electrical resistivity was estimated to be 5×10^{11} Ω cm. It is worth noting that the resistivity of our earlier InI crystals (with no material purification) was about 10^{10} Ω cm, occasionally reaching 10^{11} Ω cm. Thus the resistivity of the InI crystals was increased by a factor of 5 to 10 due to the starting material purification, and further improvement is anticipated upon implementation of more rigorous purification efforts.

The charge-transport parameters such as the mobility-lifetime ($\mu\tau$) product of electrons, and the electron mobility (μ) as well as lifetime (τ) were also estimated. Direct detection of low-energy X-rays such as 22 keV photons emitted by a ^{109}Cd source was accomplished using InI detectors as shown in Fig. 1. By studying the variation of the photopeak position as a function of the applied bias and performing Hecht analysis of the collected data, we estimated the $(\mu\tau)_e$ to be about 10^{-4} cm^2/V [6,7]. The electron $\mu\tau$ is high enough to allow efficient collection of electrons at reasonably high operating voltages. Separate estimation of the electron mobility and lifetime was performed by recording the amplitude versus time data (using a fast digital storage oscilloscope) for individual X-ray-induced pulses and then performing transient Hecht analysis [7,8]. The electron mobility was measured to be 22 cm^2/V s and its lifetime was found to be 4 μs. The mobility-lifetime product of holes was about 1 order of magnitude smaller than electrons.

The energy required to form an electron–hole pair (E_{pair}) in InI was also estimated from the direct X-ray detection measurements. The experiment involved operating an InI detector at almost 100% charge collection and recording the 22 keV photopeak (^{109}Cd source) position. Similar measurements were then performed using a silicon detector. From the relative peak positions for silicon and InI detectors and knowing the E_{pair} of silicon (3.6 eV), we estimated the E_{pair} of InI to be 4 eV.

In addition to these electrical properties, the optical quantum efficiency of InI detectors was also measured. A Bausch and Lomb monochromator (model #2D126) was used and the output intensity of the light source (tungsten "ELH" lamp) to the monochromator was measured with a calibrated silicon diode. The response of InI photodetectors to the same source spectrum was measured next, and this response was normalized with the known lamp intensity to compute the quantum efficiency versus wavelength behaviour of InI detectors as shown in Fig. 2. The quantum efficiency of InI detectors was found to be remarkably high in the 300 to 600 nm wavelength region ($>70\%$). This result is important because almost all established inorganic scintillators such as CsI(Tl), NaI(Tl), and BGO as well as promising new ones such as LSO (Lu_2SiO_5:Ce) and LuAP ($LuAlO_3$:Ce) emit in this region, and, hence, InI photo-

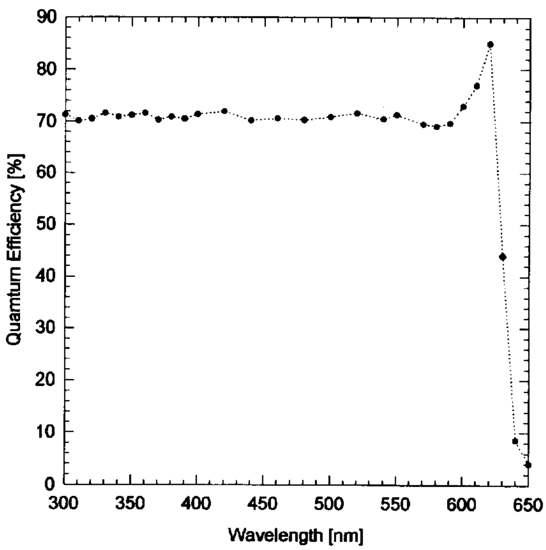

Fig. 2. Absolute quantum efficiency versus wavelength for an InI photodetector.

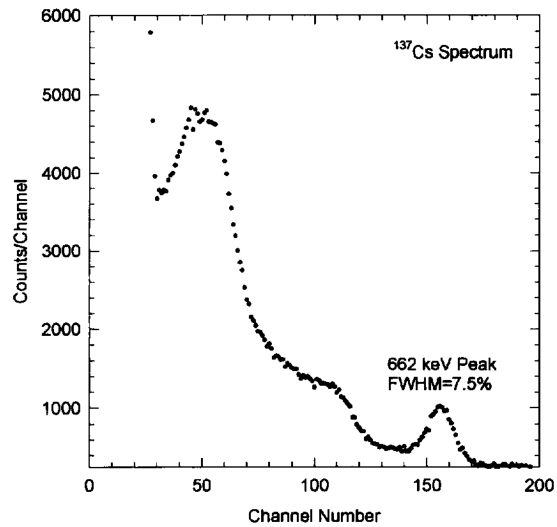

Fig. 3. ^{137}Cs spectrum taken with an InI photodetector (5×5 mm^2, 500 μm thick) coupled to a CsI(Tl) scintillator ($3 \times 3 \times 3$ mm^3). The detector was operated at 250 V bias and 2 μs integration time.

detectors would show good photoresponses with all these scintillators.

4. Scintillation studies

InI photodetectors were coupled to inorganic scintillators such as CsI(Tl) and LSO and tested as γ-ray spectrometers at room temperature. The scintillation detection tests were made by connecting the detectors to an AC-coupled charge sensitive preamplifier (Tennelec #TC170). A Canberra spectroscopy amplifier (#2020) was used and the pulse-height spectra were recorded on a Nucleus PCA-1000 board residing in an IBM compatible personal computer.

An InI photodetector coupled to a CsI(Tl) scintillator was irradiated with isotopic sources such as ^{137}Cs and ^{22}Na and the resulting γ-ray spectra are shown in Fig. 3 and Fig. 4. The InI photodetector was 5×5 mm^2 in area, 500 μm thick and was operated at a bias of 250 V. The CsI(Tl) scintillator was $3 \times 3 \times 3$ mm^3. The resolution of the 662 keV peak was measured to be 7.5% (FWHM) at room temperature, and upon calibration of the energy scale in electrons (using direct X-ray detection measurements), the peak position for 662 keV corresponded to a 15 700 e$^-$ signal. Thus, the overall light collection efficiency is about 44% which is lower than the InI quantum efficiency of 70% at the emission wavelength of CsI(Tl) ($\lambda_{max} = 540$ nm). Thus, improvement in resolution is expected as scintillator–photodetector coupling is optimized. The resolution of the 511 keV photopeak was measured to be 9.8% (FWHM), with the photopeak corresponding to a 12 000 e$^-$ signal.

The same detector was also coupled to an LSO scintillator ($4 \times 4 \times 4$ mm^3) and the ^{22}Na γ-ray spectrum was acquired as shown in Fig. 5. The resolution of the 511 keV photopeak was measured to be 14% (FWHM), and the 511 keV photopeak corresponded to a 6100 e$^-$ signal. The

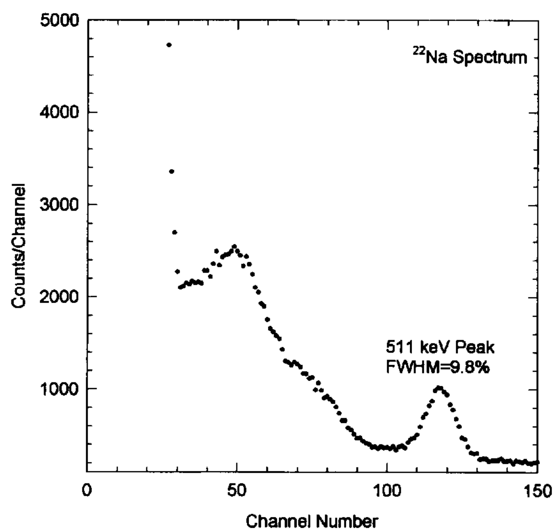

Fig. 4. ^{22}Na spectrum taken with an InI photodetector (5×5 mm^2, 500 μm thick) coupled to a CsI(Tl) scintillator ($3 \times 3 \times 3$ mm^3). The detector was operated at 250 V bias and 2 μs integration time.

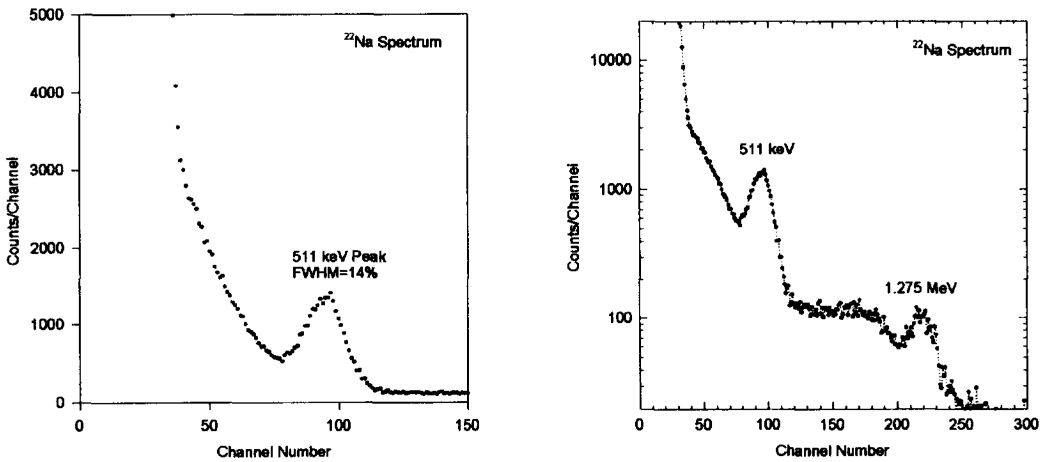

Fig. 5. ^{22}Na spectrum taken with an InI detector (5×5 mm^2, 500 μm thick) coupled to an LSO scintillator (4×4×4 mm^3), operating at 250 V bias and 2 μs integration time. The data is displayed in linear as well as logarithmic fashion, the latter also showing the 1.27 MeV photopeak, characteristic of the isotope used.

detector electronic noise was estimated to be about 700 e$^-$ (FWHM) using a test pulse. Successful detection of ^{22}Na spectra using BGO scintillators coupled to individual InI photodetectors as well as arrays has also been accomplished and will be reported in a separate publication.

5. Noise analysis

The scintillation detection experiments indicated that the electronic noise in InI photodetectors was the dominant source of resolution broadening. Hence, the electronic noise behaviour of the InI detectors was investigated to determine the magnitude of various noise components in the detectors. Electronic noise in semiconductor detectors has described in detail in the literature [9,10], and we used these existing noise models to analyze the electronic noise in InI detectors. The electronic noise is expected to arise from the following sources in InI detectors: (i) the *parallel thermal* noise due to the detector leakage current (also commonly referred to as *shot* noise), (ii) the *series thermal* noise generated in the channel of the input JFET of the pre-amplifier, and (iii) $1/f$ noise in the detector–preamplifier assembly. Expressions quantifying these noise components are listed below, and the total noise is then a quadratic sum of the individual noise components.

$$\mathrm{ENC_p} = \frac{e}{2}\sqrt{\frac{\tau I_\mathrm{d}}{q}}$$

shot noise or the parallel thermal noise from the detector current

$$\mathrm{ENC_s} = \frac{e}{q} C_\mathrm{in} \sqrt{\frac{kT}{2\tau}\left[\frac{0.7}{g_\mathrm{m}}\right]}$$

series thermal noise from the input JFET channel

$$\mathrm{ENC_f} = \frac{e}{q} C_\mathrm{in} \sqrt{\frac{A_\mathrm{f}}{2}}$$

$1/f'$ noise from the detector

$$\mathrm{ENC_t} = \sqrt{\mathrm{ENC_p^2} + \mathrm{ENC_s^2} + \mathrm{ENC_f^2}}$$

total *measured* noise

where ENC is in electrons rms, τ is the integration time of the shaping amplifier (assumed in these equations to be a simple RC–CR differentiator and integrator), I_d is the detector leakage current, g_m is the transconductance of the input JFET, k is Boltzmann's constant, q is the electronic charge, e is 2.718.., C_in is the input capacitance of the preamplifier (sum of the detector, gate and stray capacitances), and A_f is the $1/f$ spectral noise density. The additional noise components associated with the feedback and the bias resistors in the pre-amplifier design were ignored due to their negligible noise contribution when compared with the thermal noise due to the detector leakage current.

In order to estimate the noise contribution of the individual noise sources, the total electronic noise in the InI detector was measured as a function of the amplifier integration time (τ) using a test pulser. The data were then fitted to the noise model described above and estimation of the various noise sources was accomplished using a computer program. The results of such an analysis for an InI detector (5×5 mm^2 area, 0.5 mm thick) operating at 250 V is shown in Fig. 6. As seen in the figure, the shot noise (due to the InI dark current of about 2.2 nA) dominates the total electronic noise for an amplifier integration time exceeding 1 μs and all 3 sources are roughly similar in magnitude at the total noise minima of 285 e$^-$ (rms) occurring at 1 μs integration time. The series

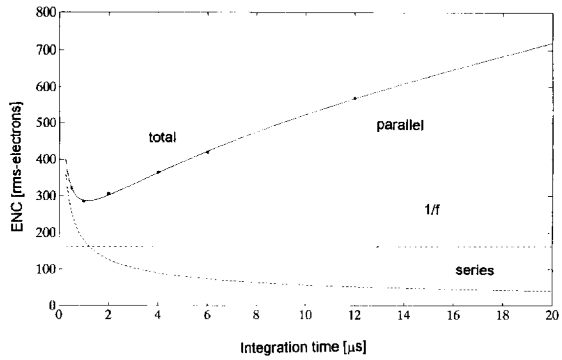

Fig. 6. Noise analysis for an InI detector (5×5 mm^2, 500 μm thick) operating at 250 V. The circles represent experimentally measured noise values, while the solid lines represent the shot (or parallel) noise, series noise, $1/f$ noise and the quadratic sum of these sources that were estimated based on the noise model to fit the experimental data. A dark leakage current value of 2.2 nA and a detector capacitance of 8.5 pF were used to estimate the parallel and series noise sources.

thermal noise (due to the detector capacitance of about 8 pF and FET gate capacitance of 2.5 pF) dominates at the smaller integration times. Thus, minimization of the detector leakage current would reduce the total electronic noise at longer integration times, which is relevant for scintillation spectroscopy using CsI(Tl), because CsI(Tl) requires an integration time of about 2–4 μs (due to its slow decay time). For use with faster scintillators such as LSO, reduction in the InI detector capacitance would enable low-noise operation at faster integration times (0.25–0.5 μs).

6. Summary

Indium iodide is a promising detector material for scintillation spectroscopy due to its high quantum efficiency in the 300–600 nm wavelength region. Progress has been made in the InI detector performance due to improved material processing. Specifically, zone refining of the starting material has resulted in an increase of InI electrical resistivity by a factor in the range 5–10. These InI crystals have also shown some improvement in the mobility-lifetime product of electrons. Photodetectors fabricated from such crystals have been successfully operated as scintillation spectrometers at room temperature and have demonstrated energy resolutions of 7.5% and 9.8%, respectively, for 662 keV and 511 keV γ-rays when coupled to CsI(Tl) scintillators. Detection of the scintillation light emitted by LSO has been achieved with InI photodetectors. Finally, evaluation of the noise characteristics of the InI photodetectors has also been performed.

Acknowledgments

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